Optical properties of beryllium-doped GaSb epilayers grown on GaAs substrate

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Abstract

In this work, the effects of p-type beryllium (Be) doping on the optical properties of GaSb epilayers grown on GaAs substrate by Molecular Beam Epitaxy (MBE) have been studied. Temperature- and excitation power-dependent photoluminescence (PL) measurements were performed on both nominally undoped and intentionally Be-doped GaSb layers. Clear PL emissions are observable even at the temperature of 270 K from both layers, indicating the high material quality. In the Be-doped GaSb layer, the transition energies of main PL features exhibit red-shift up to ~7 meV, and the peak widths characterized by Full-Width-at-Half-Maximum (FWHM) also decrease. In addition, analysis on the PL integrated intensity in the Be-doped sample reveals a gain of emission signal, as well as a larger carrier thermal activation energy. These distinctive PL behaviors identified in the Be-doped GaSb layer suggest that the residual compressive strain is effectively relaxed in the epilayer, due possibly to the reduction of dislocation density in the GaSb layer with the intentional incorporation of Be dopants. Our results confirm the role of Be as a promising dopant in the improvement of crystalline quality in GaSb, which is a crucial factor for growth and fabrication of high quality strain-free GaSb-based devices on foreign substrates.

Keywords: Gallium Antimonite, Beryllium doping, photoluminescence, threading dislocation, strain

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I. INTRODUCTION

GaSb-based III-V semiconductors system has attracted extensive research and commercial attentions in the past three decades. High performance semiconductor devices such as field-effect transistors [1], lasers diodes [2] and photodetectors operating at mid- to long-wave infrared regimes [3-5] have been demonstrated based on the InAs/GaSb/AlSb alloys and heterostructures. Ideally, epitaxial growth of GaSb layers are performed on the native lattice-matched GaSb substrates. However, cost of the GaSb substrates is relatively high, and the free carrier absorption of the substrate in the mid-infrared region hinders the further application of back-side illumination for the devices [6]. In addition, integration of GaSb substrate-based devices with read-out circuit is hard to implement using the monolithic technology [7]. Thus, for both research and commercial interests, epitaxial growth of GaSb on low-cost GaAs substrate is highly motivated. Nevertheless, the utmost challenge is the large lattice mismatch between GaSb and GaAs (~ 7.8 %), which could generate a large amount of defects and affect the electrical and optical properties of the devices.

In order to overcome the material problems associated with the large lattice mismatch, insertion of various buffer layers have been utilized, such as the AlSb/GaSb superlattices [8, 9] and AlGaAsSb metamorphic layers [10]. These methods have been demonstrated to reduce the density of dislocations and improve the quality of the GaSb/GaAs heterostructure. Alternatively, intentional incorporation of impurities into the material layer has also been implemented to suppress the large density of defects. For instance, improvements in surface morphology and transport properties by beryllium (Be) doping were reported in an InAlAs-based high-electron-mobility transistor grown on GaAs [11]. Moreover, Gutiérrez et al. [12] have studied the joint influences of low growth temperature and Be doping on the behaviors of threading dislocation in GaSb layer grown on GaAs substrate. They observed a seven times reduction in the density of threading dislocations in the Be-doped GaSb layer, which was explained by the effective "pinning" of the dislocations by the Be dopants, i.e., the Be-related hardening mechanism. Nevertheless, previous studies have focused on the effect of Be doping in crystalline quality and electrical properties of GaSb, but the impact on the optical properties of GaSb is still lacking, which could be a crucial material issue in the design and characterization of GaSb-based optoelectronic devices.

In this paper, we investigate the effects of Be doping on the optical properties of GaSb epilayers grown on GaAs substrate by temperature- and excitation power-dependent photoluminescence. Distinguishable improvements in the PL properties are identified in the GaSb layer with intentional Be doping when compared with that of the undoped GaSb layer. These results provide direct optical evidences of strain relaxation in GaSb due possibly to the reduction of threading dislocation propagated into the active layer.

II. EXPERIMENTAL DETAILS

Nominally undoped and intentionally doped GaSb layers with Be (hereafter referred to as "GaSb:Be") were grown on GaAs (001) substrates by molecular beam epitaxy (MBE). Details of the samples growth procedures have been reported elsewhere [12]. The schematic layout of the sample structure is shown in Table 1. Prior to the growth of the GaSb epilayer, a 200 nm undoped GaAs buffer layer was grown on top of the GaAs substrate at 590 ° C. For the intentional Be-doped sample, the doping concentration was 1×10^{19} cm^{-3}. The thicknesses of both undoped GaSb and GaSb:Be samples were 50 nm.

For the photoluminescence (PL) measurement, the samples were mounted in a close-cycled He cryostat with temperature varied from 9 K to 300 K [13]. The samples were optically excited by the 532 nm line of a laser. The luminescence signals were collected by a Fourier transform infrared spectrometer in continuous- rather than step-scan mode, in conjunction with a room-temperature InGaAs photodetector [14, 15]. In order to investigate the excitation-power dependence of PL, a laser-power controller was used such that the incident power were tuned from 0.5 mW to 95 mW [15].

Table 1. Schematic layout of the GaSb/GaAs heterostructure.

Active	GaSb	50 nm	Undoped / 1×10^{19} cm^{-3}
Buffer	GaAs	200 nm	Undoped
Substrate	GaAs		

III. RESULTS AND DISCUSSIONS

Temperature-dependent PL spectra measured from the undoped GaSb and GaSb:Be samples are shown in Fig. 1. Throughout the measurement the excitation laser power was maintained at 50 mW for both samples. At the temperature of 9 K, four PL emission features can be clearly identified. For the undoped GaSb sample, these peaks are found at: P_1 (0.959 eV), P_2 (1.142 eV), P_3 (1.319 eV) and P_4 (1.485 eV), respectively. This PL peak energy range was reported only in GaSb-based nanostructures grown on GaAs substrate, such as quantum rings and quantum dots [16, 17], and it is higher than the PL energy in most of the GaSb epilayers grown on GaAs reported previously (0.71-0.83 eV), [8, 9, 18-24] regardless of the growth techniques and doping condition. We notice that the thickness of GaSb layers investigated in those reports range from 1 - 20 μm, which is significantly larger than that of the samples studied in the current work, i.e. 50 nm. Considering the relatively large lattice mismatch in the GaSb/GaAs heterostructure (~7.8 %), we therefore surmise the blue-shift of PL features in our samples is due to the not fully-relaxed strain existed in the thin GaSb layers caused by the propagation of a large number of dislocations generated at the GaSb/GaAs interface [21]. To verify this assumption, the band-edge energies of the GaSb layer grown on GaAs in both fully strained and fully relaxed conditions at different temperature were calculated based on the material parameters provide by Vurgaftman et al. [25], as shown in Table 2. At temperature of 9 K, the band-edge energy of fully strained GaSb layer is 1.465 eV, whereas the band-edge of fully relaxed layer is at 0.812 eV, consistent with the low temperature band-gap values generally reported for GaSb. It is expected that the blue-shift of the PL peak energies would be reduced by growing thicker layers in the future work [21].

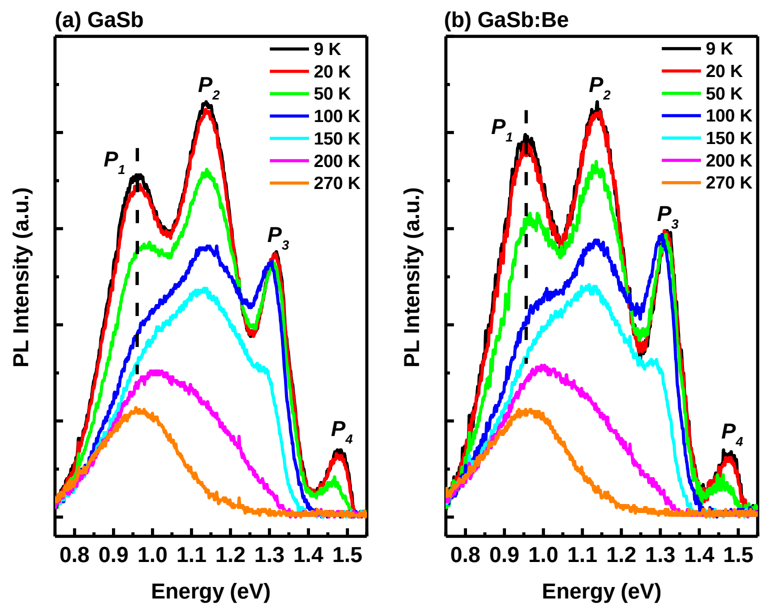

Fig. 1. Temperature-dependent PL spectra measured from (a) undoped GaSb and (b) GaSb:Be layers. The excitation laser power was maintained at 50 mW throughout the measurement.

Table. 2. Estimated band-gap energy for GaSb layer grown on GaAs substrate at different temperature.

Temperature (K)	Fully relaxed GaSb E_g (eV)	Fully strained GaSb E_g (eV)
9	0.8117	1.4654
77	0.7989	1.4531
100	0.7925	1.4469
200	0.7606	1.4157
270	0.736	1.3919
300	0.725	1.3815

As can be seen in Fig. 1, PL spectra of two samples display similar temperature dependences. When temperature increases from 9 K, the energy of peak P_1 displays a clear blue-shift in both samples, and the PL signal quenches rapidly compared with other features. It disappears completely after temperature of 150 K. For peak P_2, no clear energy shift is observed until 100 K, and the peak shifts monotonously to lower energy with temperature beyond 100 K. Peak P_3 has much lower PL intensity compared with the former two, and it also shifts to lower energy with increasing temperature; however, its PL intensity quenches much slower than that of P_1 and P_2, in which at 100 K it dominates the PL spectra simultaneously with P_2, but drops quickly when temperature increases to 150 K. Finally the weakest PL feature is seen at P_4. It exhibits red-shift with increasing temperature and it completely quenches after 50 K. It is noted that after 150 K the spectra from both samples transform to a single broad luminescence band, which is peak P_2. From previous temperature-dependent PL studies on GaSb layers, it was found that at a temperature typically higher than 60 K, the PL feature comprises only a single board band, which was attributed to band-to-band transition [6, 18, 24, 26], i.e. recombination of free electrons and holes. In addition, in an attempt to explain the origin of various PL peaks involving p-type acceptor impurities, Agert et al. [26] proposed an energy scheme model in moderately to highly doped GaSb which considered local variation of band-edge due to the large scale fluctuation of the impurity concentration in the layer. The consequence of this model is that the emission peak with highest energy and dominates the PL spectra at high temperature regime should be interpreted as the band-to-band transition. Thus, we could assume the transition responsible for the peak P_2 in both undoped and Be-doped GaSb sample is a band-to-band recombination process in the top GaSb layer. Nonetheless, from Fig. 1 it is clear that the PL energies of peak P_3 and P_4 are higher than that of P_2. In order to reconcile our PL spectral result and the band-edge model proposed by Agert et al., we notice that the absorption coefficient of GaSb under an excitation wavelength of 532 nm is estimated to be ~ 4.35×10^5 cm^{-1} [27] , which corresponds to an optical penetration depth of ~ 23 nm, i.e., about the same order as the samples thickness. It is also noted that in high quality GaSb epillayers (with a high Hall mobility) grown on GaAs, the diffusion length of photo-generated carriers as long as ~1 μm have been reported [19]. Thus, for our samples with thickness of 50 nm, as a consequence of excitation light penetration and carrier diffusion, the PL emissions observed at P_3 and P_4 could possibly be originated from the sample regions which are close to the GaSb/GaAs interface or in the buffer layer. PL emission occurred here would be affected by the large compressive strain caused by the interface lattice mismatch, as evidenced by the much lower PL efficiency and higher energy observed from peak P_3 and P_4 in Fig. 1. In particular, in view of the emission energy and the very weak signal of peak P_4, we attribute this PL emission to the carrier radiative recombination in the GaAs buffer layer. The following discussion will be focused on peak P_1, P_2 and P_3 which are considered to be associated with the GaSb layer and GaSb/GaAs interface.

Fig. 2 shows the temperature dependences of emission energy for the three PL peaks at 9 - 50 K extracted from Fig. 1. For the undoped GaSb layer (GaSb:Be), the energy of peak P_1 shifts from 0.959 eV (0.954 eV) at 9 K to 1.017 eV (1.014 eV) at 150 K, exhibits a total shift of ~ 60 meV. The blue-shift of PL transition energy with increasing temperature is commonly seen in many III-V semiconductors where localization of electrons is present, typically caused by the band-tail states due to spontaneous group-III sublattice ordering or impurities fluctuation [28]. Such temperature induced blue-shift were also seen in the low temperature PL peaks at ~ 0.777 eV (label as "A line") and ~ 0.8 eV (label as "E line") in GaSb layers [6, 24, 26]. The carrier process ascribed to these two peaks are either donor-acceptor pair (DAP) recombination, or from localized electron to neutral acceptors by considering the band-edge fluctuation. As seen from Fig. 2, the PL energies emerged from the GaSb:Be sample are consistently lower than that of the undoped sample for the three emission peaks. Despite the small energy difference for peak P_3 (possible reason will be discussed later), a red-shift up to ~ 7 meV (peak P_1, at 50 K) can be seen in the PL peak energies of the GaSb:Be sample. In addition, the peak width of the PL emission, which is expressed as the Full-Width-at-Half-Maximum (FWHM), is presented in Fig. 3. As expected the PL bands become broader with increasing temperature due to the enhanced phonon scattering, and the FWHM of the PL bands from the GaSb:Be are smaller than that of the undoped sample. The red-shift of PL emission energy as well as the decrease of emission linewidth observed in the GaSb:Be sample jointly suggest a reduction of residual compressive strain in the Be-doped GaSb layer due to the reduction of dislocation density [11]. When Be dopants are introduced in the GaSb sublattice, the density of threading dislocation prorogated into the top GaSb layer from the lattice mismatched GaSb/GaAs interface is reduced due to the Be-related defect "pinning" mechanism , which has been evidenced by high-resolution transmission electron microscopy in Ref. [12].

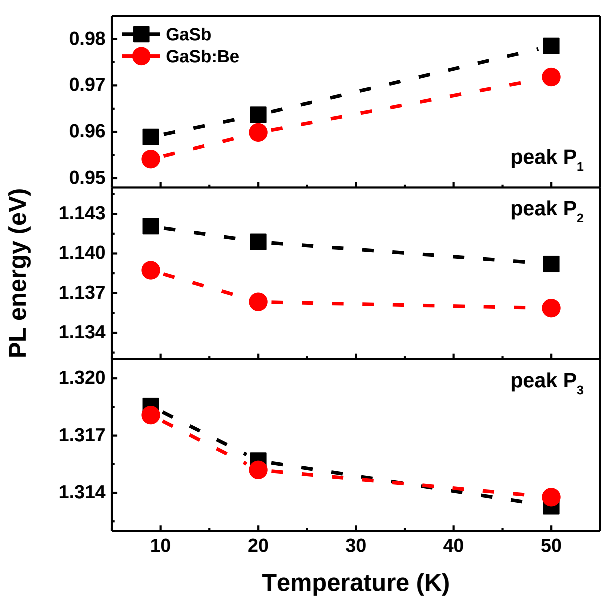

Fig. 2. Temperature versus PL peak energy measured from both samples at 9 - 50 K.

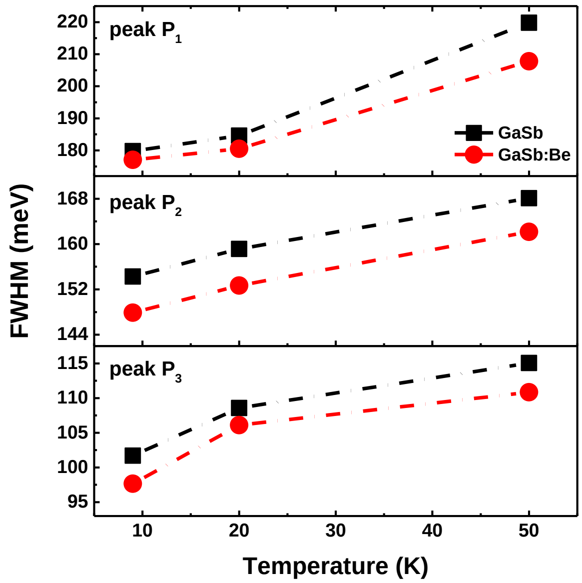

Fig. 3. Temperature versus PL peak width (FWHM) measured from both samples at 9 - 50 K.

To further study the effect of Be dopants on the optical quality of the GaSb layers, four representative PL spectra measured from the two samples are plotted in Fig. 4. At the lowest temperature of 9 K, increase in PL intensity can be observed for both peak P_1 and P_3 in the GaSb:Be sample, whereas for the peak P_2 and P_4 the effect of intentional Be doping on PL intensity gain is not evidenced, which confirm the irrelevance of peak P_2 and P_4 to impurity-related recombination as discussed above . Due to the fast temperature quenching nature, the PL intensity increase of P_1 in GaSb:Be sample slows down at 100 K, and no PL intensity gain is observable when temperature is further raised above 150 K. In an attempt to clarify the PL thermal quenching mechanism for each emission feature, the experimental PL intensity is fitted by the Arrhenius formula involving one nonradiative recombination channel [13]:

$$I(T) = \frac{I(T_0)}{1 + a_0 T \exp(-E_a / k_B T)} \tag{1}$$

where $I(T_0)$ is the integrated intensity at a reference low temperature (9 K in this case), a_0 is a constant associated with the radiative and nonradiative recombination probability, E_a is the activation energy, and k_B is the Boltzmann's constant. The Arrhenius plots for both samples are shown in Fig. 5. Very good fits are obtained for both samples, which enable the extraction of activation energy. The activation energies for peak P_1, P_2 and P_3 are 2.3 meV, 12.1 meV and 46.2 meV respectively. With the incorporation of Be doping, the activation energies increase for all PL features. In particular, a remarkable increase of 10 meV is seen in peak P_3. Since the various dislocation and defects could serve as nonradiative recombination sites in the GaSb layer [26], the gain in PL signal and thermal activation energy in the GaSb:Be sample further confirm the effective reduction of dislocation density as a result of the intentional Be doping. It is noteworthy to mention that improvement of PL efficiency have also been reported in GaSb layers grown on both GaSb and GaAs substrates with intentional Si and C doping [20, 24, 26]. Although Si behaves as an amphoteric dopant in III-V materials, in GaSb it tends to integrate as an acceptor in the Sb anionic sublattice via atomic substitution, similar to the Be dopants in GaSb.

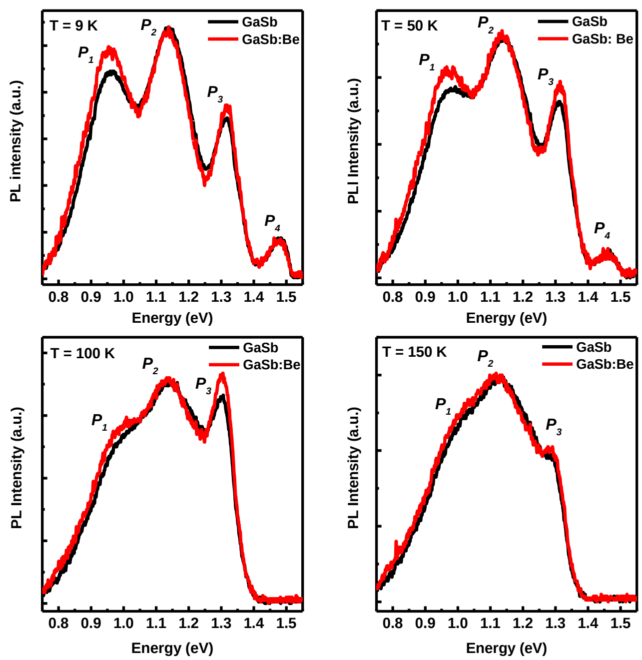

Fig. 4. Four representative PL spectra measured from both samples at 9 - 150 K.

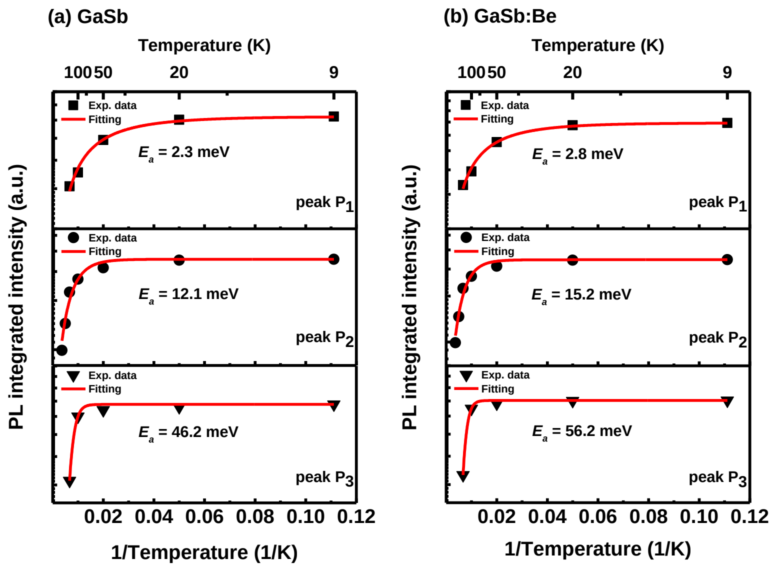

Fig. 5. Arrhenius plot of the integrated intensity of the three main PL peaks for (a) undoped GaSb and (b) GaSb:Be layers. The symbols represent experimental data and the solid lines are the fitting curves by using Eq. (1).

Finally, in order to unveil the origin of peak P_1 and P_3, excitation power-dependent PL measurements were taken on the two samples, as depicted in Fig. 6. During the measurement the excitation power was varied from 0.5 mW to 95 mW, while the temperature was kept at 9 K. At low excitation power, only three emission peaks of P_1, P_2 and P_3 can be observed, and the PL signal at P_4 becomes merely distinguishable when the laser power rises to 5 mW. With further increase of excitation power, except the overall gain in intensity of all PL features, no obvious shift in peak position can be identified, including peak P_1. This helps to exclude the possible attribution of P_1 as a DAP recombination, since the hallmark of a DAP process is the remarkable blueshift of peak energy with increasing excitation intensity [21]. Considering the rapid thermal quenching and the small activation energy of < 3 meV obtained from peak P_1, we thus assign this peak to the transition from the electrons localized at the conduction band-tail states to a shallow acceptor. It is well known that the background conductivity in unintentionally doped GaSb is always p-type, regardless of the growth techniques and doping condition. The defect state responsible for this behavior is a native acceptor (NA), which might be a point defect complex composed of Ga vacancy (V_{Ga}), Sb vacancy (V_{Sb}), or a Ga-on-Sb antisite (Ga_{Sb}) with both singly and doubly ionizable nature [7, 8, 21-24, 26]. When temperature increases, electrons localized at the band-tail states are thermally excited to the higher tail states, which leads to the initial blue-shift of PL emission peak before the temperature-induced band-edge shrinkage starts to dominate. At elevated temperature, the shallow acceptors are thermally decomposed due to the very small activation energy (< 3 meV), causing the emission at peak P_1 quenches rapidly as observed in Fig. 1. For peak P_3, the higher emission energy than the band-to-band recombination (peak P_2) and much weaker intensity advice that this PL process may take place between free electrons and the native acceptors at the region close to the GaSb/GaAs interface. One important point to be noted is the large activation energy (E_a) obtained from this PL process. As shown in Fig. 5, for the undoped GaSb layer, E_a for peak P_3 is ~ 46 meV, which is slightly larger than the reported range of 20 - 40 meV for the NA acceptors if single ionizable state is assumed [18, 19, 24]. The activation energy increases to ~ 56 meV with the incorporation of Be. This value is in very good agreement with the Be thermal activation energy of 55 meV extracted from the temperature-dependent Hall measurement conducted on Be-doped GaSb layer grown on GaAs by MBE [29]. This result further confirms the effective integration of Be in the NA acceptor sites, possibly by occupying the various V_{Ga} and V_{Sb} vacancies. Therefore, the transition at peak P_3 in the GaSb:Be sample could be due to the recombination of free electrons to the Be acceptor states which takes place at the GaSb/GaAs interface region. The large density of unrelaxed strain in the interface may be the cause for the much smaller PL energy red-shift of P_3 between the two samples, as observed in Figure 2. We also observe similar PL thermal activation energy of ~59 meV in undoped GaSb layer grown on GaAs substrate with the insertion of AlSb and GaSb buffer layers [8]. The presence of AlSb/GaSb buffer layers have been demonstrated to restrict the threading dislocations in GaSb layer [9], thus we believe the intentional doping of Be could serve as a promising alternative for achieving material quality improvement in GaSb layers grown on lattice-mismatched substrates.

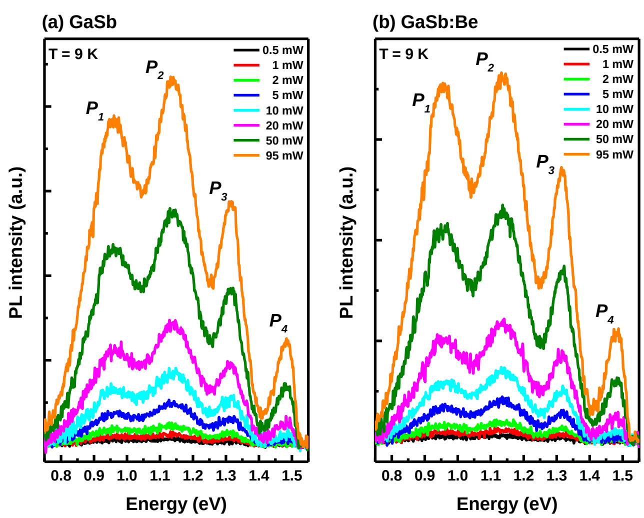

Fig. 6. PL spectra measured as a function of excitation power from (a) undoped GaSb and (b) GaSb:Be layers. All spectra were recorded at the same temperature of 9 K.

IV. CONCLUSION

In summary, the effect of p-type Be doping on the optical properties of GaSb epilayer grown on lattice-mismatched GaAs substrates have been investigated by temperature- and excitation power-dependent PL measurements. Efficient PL emission was observable even at temperature of 270 K, indicating the high material quality despite the existence of large lattice mismatch. We show that in the Be-doped sample, the PL peak energies exhibit a red-shift up to ~7 meV, and the linewidths also decrease. The acceptors-related PL transition demonstrated a gain in PL intensity, and a carrier thermal activation energy of ~ 56 meV was obtained. These results suggest an improved optical quality in the Be-doped GaSb layer, which is due probably to the reduction of threading dislocation propagated into the active layer from the strained GaSb/GaAs interface. Our results confirm the potential of Be as a promising p-type dopants for the development of high quality GaSb-based optoelectronic devices grown on low-cost, yet lattice-mismatched foreign substrates, such as GaAs and Si.

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